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DE89 016588

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TITLE

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SUBMITTED TO

The 5th International Symposium on Polarization and Correlation in Electric and Atomic Collisions, Stevens Institute of Technology, Hoboken, N.J., August 2-4, 1989.

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THE EFFECTS OF A FINITE SCATTERING VOLUME ON THE DETERMINATION OF ELECTRON IMPACT COHERENCE PARAMETERS

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1. Introduction

In electron-atom beam-beam scattering experiments, the interaction region defined by the intersecting beams and the viewcone of the detector should be small in comparison with other characteristic dimensions of the scattering geometry and the energy and angular resolution of the apparatus should be narrow with respect to the ranges over which quantities of interest undergo significant changes. In this ideal case, the scattering signal can be assumed to originate from a point-like source and the data obtained from the measurements can be assigned to well-defined electron impact energy (E_o) and scattering angle (θ_e). In practice, the dimensions of the scattering volume and the energy and angular resolution of the apparatus are always finite and a rigorous treatment of the scattering data should take this into account. In conventional electron scattering measurements of the differential scattering cross section (DCS) these considerations, in general, cause no serious problems. The DCS can then be associated with nominal scattering angles and impact energies and represents a sum over final and average over initial experimentally indistinguishable processes. A discussion of these matters in electron scattering DCS measurements has been given, for example, by Brinkmann and Trajmar[1]

In recent years, a large body of data has accumulated in the field of electron impact induced orientation and alignment of atomic valence shells[2]. Coherence and correlation experiments used to examine these aspects of the collision process represent a considerable refinement over conventional DCS measurements. In principle, only the scattering signal originating from an ensemble of excited atoms prepared in a well-defined quantum mechanical state is measured. The selection of this ensemble is carried out either by coincidence detection of the inelastically scattered electron and emitted photon, or by laser preparation of the excited state atomic target and detection of the superelastically scattered electron. In both cases, a photon incidence vector and polarization vector are defined whose directions with respect to the scattering plane must be well-specified in order that unambiguous conclusions can be drawn about collision induced alignment and orientation (or, equivalently, about the electron impact coherence parameters, EICP). A rigorous evaluation of scattering data from coherence and correlation experiments should, therefore, include the convolution of the scattering angle as well as the photon direction and polarization angles with the finite angular resolution of the apparatus. Generally, however, the picture of an ideal point-like scattering has been applied to the interpretation of coherence and correlation data.

A recent study by Martus *et al.*[3] does take into account the effect of averaging over the finite range of unresolved scattering angles observed by the electron detector in a measurement of the P1 polarization correlation parameter. Here, we describe a more rigorous approach to the problem of determining the influence of a finite scattering volume in electron-photon coincidence experiments and in measurements of superelastic scattering from laser-excited atoms. We treat the interaction volume as an ensemble of individual scattering points to each of which is attached a collision coordinate frame that may differ significantly from a laboratory fixed coordinate frame. A "true" scattering plane, defined by the incident and outgoing electron momentum vectors of a collision event occurring somewhere within the ensemble can be quite different from the "nominal" scattering plane defined in the laboratory frame. The transformation of coordinate frames has extremely important consequences for the measurement of EICP and is unaccounted for by averaging over the scattering angles alone. Photon incidence and polarization vectors which are specified in the laboratory coordinate frame by spherical angles θ_l and ϕ_l and by polarization angle ψ_l are given in the collision coordinate frame by angles θ_c, ϕ_c and ψ_c

which can, depending on the nominal scattering angle, vary radically for collision events occurring throughout the extended scattering volume. This radical behavior is expected for nominal scattering angles close to zero since the rotation of a collision frame relative to the laboratory frame for a scattering event displaced from the "nominal" scattering plane can be severe at small scattering angles. In this situation, the angular resolution of the system is no longer narrow with respect to the range over which the photon polarization vector and the photon direction vector (in particular) undergo significant variation. The extraction of EICP at these scattering angles becomes meaningless in the simple point scattering picture. Surprisingly, we have also discovered that a scattering volume of finite extent can have a dramatic influence on the measurement of EICP at scattering angles far from zero. In these [illegible]

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[illegible]

2. Model Calculations

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linearly polarized photons:

$$I \sim A + B'\cos 2\psi + B''\sin 2\psi \qquad (1)$$

where the coefficients A, B′ and B″ have been given[4,5] as functions of the EICP and the photon incidence vector direction angles θ_n, ϕ_n measured in the collision coordinate frame. Implementation of the model involves carrying out the coordinate frame transformation associated with a scattering center located at position vector $\vec{r}_j$ from the laboratory frame origin. The relation between the photon angles θ_ν, ϕ_ν, ψ_ν specified in the laboratory frame and the angles θ_n, ϕ_n, ψ in the collision frame is thereby determined as is the relation of the true scattering angle, θ_e, to the nominal scattering angle, θ_e. Fig. 1 presents a comparison between the collision frame associated with a scattering event located at position vector $\vec{r}_j$ and the collision frame associated with an "ideal" scattering event taking place at the origin of the laboratory frame ($\vec{r}_j = 0$). This latter scattering event defines the nominal scattering plane and the nominal scattering angle, θ_e. The collision frame associated with the offset scatterer is clearly rotated with respect to the laboratory frame as evidenced by the direction of the vector $\hat{Y}^{j}_{c}$ (normal to the true scattering plane) compared to $\hat{Y}_{c}$ ($\hat{Y}_{c}$ normal to the nominal scattering plane.)

Fig. 1 Electrons emitted by the source (G) are scattered into detector (D). Collision coordinate frames associated with two different scattering events taking place within the extended interaction volume are shown (see text).

In our model, a weighted average of the scattering event intensity over the distribution of scattering centers is taken according to:

$$I_\Sigma = \sum_{ij} a_{ij} I_j(\hat{k}'_j)$$

where I_Σ is the average superelastic or coincidence intensity and a_{ij} are weighting factors discussed below. The signal intensity arising from a particular collision frame is denoted $I_j(\hat{k}'_j)$ since (for an assumed parallel incident electron beam) this frame is completely specified by the displacement vector $\bar{r}_j$, the nominal scattering angle, θ_e^o and the momentum vector, $\hat{k}'_j$, of an electron scattered into the detector viewcone. The summation over index i in Eq. 2 represents an approximation to the effect of finite angular resolution in the electron detector. The contribution from all discrete scattering points chosen to represent the extended volume is calculated in the summation over index j. The weighting factors, a_{ij}, represent the effect of the spatial intensity distribution of the incident electron beam, the spatial response of the electron detector and the behavior of the DCS over the range of scattering angles defined by the extended scattering volume.

The sophistication of the model can be greatly increased to include, for example, the finite acceptance angle of the photon detector (or the divergence of the incident laser beam in the superelastic scattering case), the density distribution in the target gas beam and so on. We have found, however, that a relatively crude description of the scattering geometry can provide the essential aspects of the finite volume effect. In the results presented here, an array of 5 points, equidistantly spaced along a segment of the Y_{lab} axis, was used to represent the extended scattering volume. The effect of the detector viewcone was simulated by associating 5 scattering directions with each of the 5 scattering points in the linear array. The weighting factors, a_{ij}, reflect only the spatial intensity distribution (Gaussian) of the incident electron beam.

3. Results

3.1 Superelastic Scattering from Laser-Excited ^{138}Ba($\cdots$6s6p ^{1}P)

Efforts to measure EICP for the ^{138}Ba $^1S_0$ to $^1P_1$ transition in a superelastic scattering experiment have been reported by Register *et al.*[6] An experimental configuration was adopted in which the laser beam lay within the (nominal) scattering plane and the degree of modulation of the superelastic intensity, I_s, as a function of laser beam polarization angle, ψ_ν, was measured. During the course of their investigation, Register *et al.* discovered that this modulation, $I_s(\psi_\nu)$, was, unexpectedly, asymmetric with respect to $\psi_\nu = 0$ where $\psi_\nu = 0$ is defined to be the angle at which the laser beam polarization vector lies within the scattering plane. Furthermore, this asymmetry was scattering angle dependent in a regular way and was found to be reproducible despite major variations in experimental conditions. The origin of this asymmetry remained unexplained and EICP could not be extracted, with confidence, from these measurements. This led us to reinvestigate the cause of the asymmetry in more detail, both experimentally and theoretically, with special emphasis on the effects of a scattering volume of finite extent.

Superelastic scattering data can be straightforwardly analyzed by fitting Eq. 1 to the observed $I_s(\psi_\nu)$ modulation curves. For this purpose, Eq. 1 is written in the equivalent form:

$$I_s(\psi_\nu) \sim 1 + \eta \cos(2\psi_\nu + 2\alpha) \qquad (2)$$

where η is the modulation depth and α is the modulation phase shift.

For a laser beam lying in the scattering plane, theory[4,5] predicts that $\alpha = 0$. However, the asymmetry observed by Register *et al.* and by us, in a more recent series of experiments, can be represented by a modulation phase shift which is non-zero. Fig. 2 shows the values of η and α extracted from our recent superelastic scattering measurements and the results of the corresponding model calculations for incident electrons of 30eV impact energy. These model calculations employed the theoretical EICP of Clark *et al.*[7] which indicate that the ^{138}Ba($\cdots$6s6p ^{1}P) state is almost purely LS coupled. It is important to note that, for a purely LS coupled state, an ideal single atom collision occurring at the origin of the laboratory frame (for laser incidence angle $\phi_\nu = 0$) would give $\eta = 1$ and $\alpha = 0$ at all scattering angles. Fig. 2 illustrates that serious deviations from these predicted values occur in an actual experiment and that these deviations are explained by the extended scattering volume model.

The model calculations indicate that, for a laser beam incidence vector lying in the nominal scattering plane, the phase shift differs from zero when the extended scattering volume is asymmetrically distributed in the normal direction to the nominal scattering plane. We have also verified experimentally that the deviation of α from zero is tied directly to asymmetry in the distribution of excited-state scatterers[8,9].

The distortion in the modulation depth, however, persists regardless of how the scattering volume is distributed. These results imply that the phase shift problem could conceivably be eliminated by ensuring a symmetrically distributed scattering source. However, EICP are extracted from the modulation depth η, which suffers significant distortion whenever the scattering volume is finite in extent.

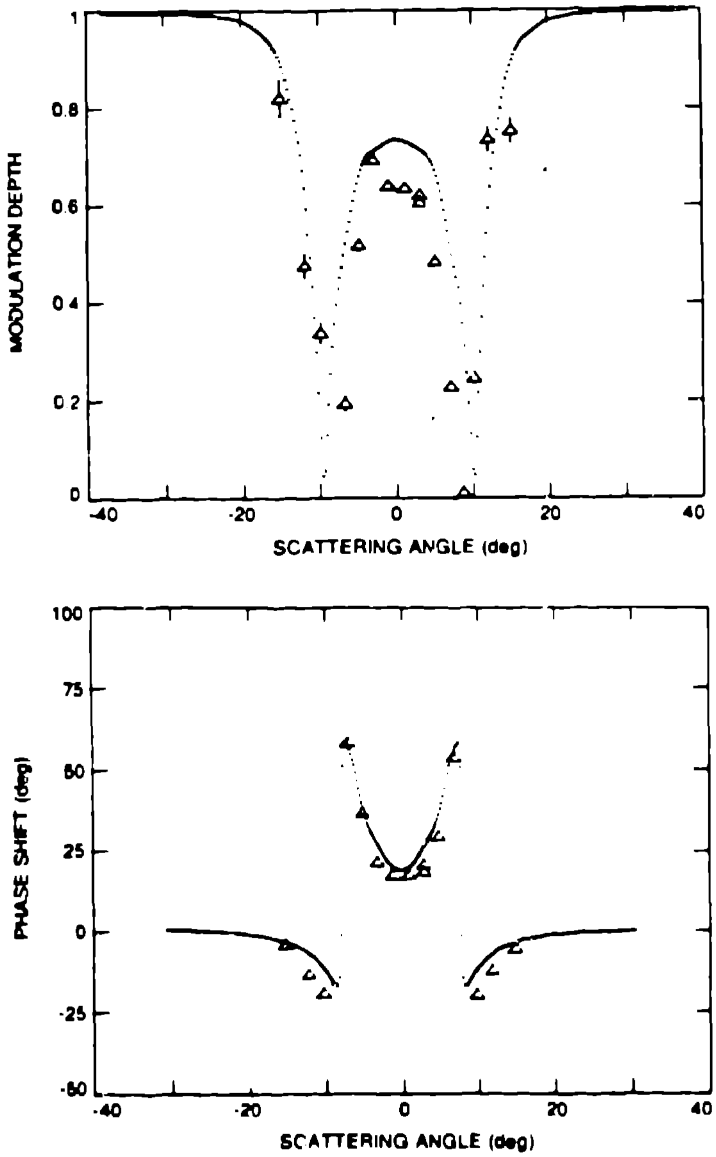

Fig. 2 Model calculations and experimental data[8] for superelastic scattering from $^{138}Ba(\cdots 6s6p\ ^1P)$ at 30 eV. Laser beam incidence angles are $\theta_\nu = 90°$; $\phi_\nu = 0°$. (a) Modulation depth vs. nominal scattering angle; (b) Phase shift vs. nominal scattering angle.

Fig. 2 also reveals that the distortion in modulation depth produces relatively well-defined features which appear at specific scattering angles. For the superelastic scattering process under discussion, the scattering angle location of these features is closely tied to the behavior of the alignment angle, γ, of the excited state charge cloud produced by the time-inverse inelastic scattering process. When this alignment angle is such that the laser photon incidence vector lies along the major axis of the charge cloud (assuming a collisionally excited "p" type orbital) then the measurement becomes extremely sensitive to extended volume effects[5]. Although it is not apparent from the figure, we have also found that the severity of the geometry-induced distortion is related directly to the shape of the charge cloud produced by the time-inverse inelastic process. In particular, when the ratio of length to width of this charge cloud is large at the "critical" alignment angle described above, then the distortion in η becomes severe. The smaller this ratio, the less sensitive the measurement becomes to extended volume effects. Of course, this ratio is given by the P_l^+ coherence parameter and we claim that, when P_l^+ is close to unity at the critical alignment angle, one can expect a strong influence of geometry on the measurement of η.

3.2 Measurements of the P4 Coherence Parameter in the Rare Gases

Conclusions about the influence of an extended scattering geometry on "laser-in-plane" superelastic scattering experiments are directly transferrable to studies concerning the P4 Stokes parameter in rare gas $J = 0$ to $J = 1$ excitations. We, therefore, extended our modelling effort to the rare gases and, in Figs. 3 through 5, the results are compared with the only P4 measurements so far available in the literature[9]. The same crude model was employed as in the case of superelastic scattering from $^{138}Ba(\cdots 6s6p\ ^1P)$. The photon detector was placed in the nominal scattering plane at 90° relative to the incident electron beam and was assumed to have infinite angular resolution. The EICP of Bartschat and Madison[10] were used.

Fig. 3 shows the P4 vs. scattering angle behavior for the ^1P state excitation in Ne. Dots (crosses) represent modelling calculations carried out with a finite scattering volume of 1mm (2mm) extent. The ^1P state is described by pure LS coupling so that P4 is expected to be unity over all scattering angles. Near zero degrees scattering angle, the influence of a finite scattering volume reduces the P4 polarization to a minimum of about 0.8.

For the heavier rare gases, Kr and Xe, we found that, as discussed above, the shape of the modelled P4 vs. scattering angle curve was quite sensitive to the behavior of the theoretical P_l^+ near the "critical" alignment angle. The model calculations plotted in Figs. 4 and 5 result from slightly modifying the P_l^+ behavior predicted by Bartschat and Madison. The calculated behavior of the alignment angle was left unmodified.

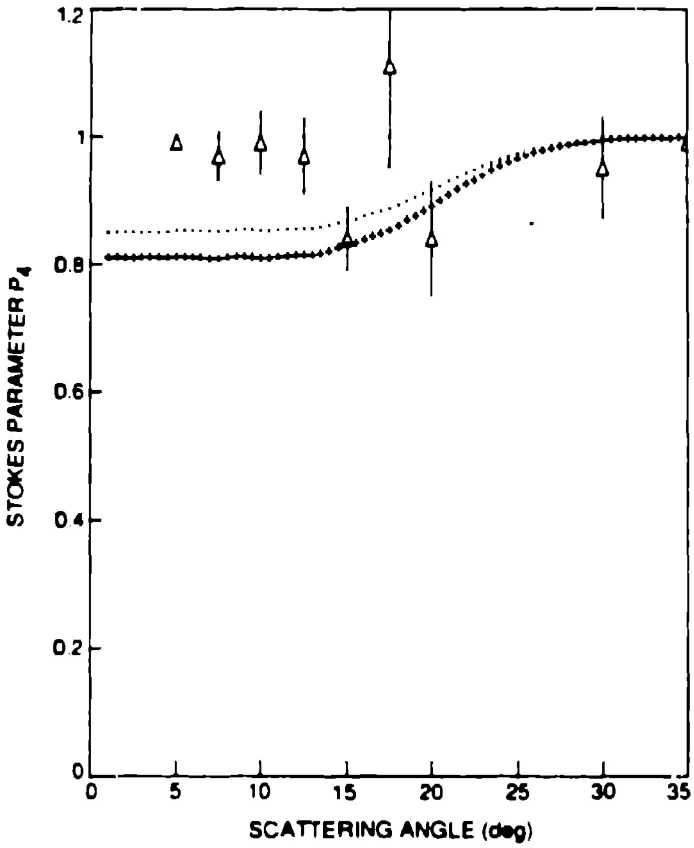

Fig. 3 Model calculations and experimental data[9] for excitation of Ne $3s'[1/2]_1^0$ $^1P_1$ at 80 eV impact energy.

We performed this exercise in order to check whether the experimental P4 data, whose deviation from unity is ascribed to spin-orbit coupling effects[9], could be fit by adjusting only the theoretically predicted behavior of P_l^+ which is not associated with spin-orbit coupling. The fit of the model results to the experimental data in Figs. 4 and 5 is reasonable, suggesting that, under conditions where spin-orbit coupling effects are negligible (i.e. $\cos\epsilon = 1$ or $\rho_{00} = 0$), extreme distortion in the P4 measurement can occur for a particular behavior of the coherence parameters P_l^+ and γ (or, equivalently, λ and χ) and a scattering volume of finite extent. The existence of an extended scattering geometry is a necessary condition for this extreme behavior in P4 to manifest itself. This is illustrated by the fact that, for an ideal, single-point

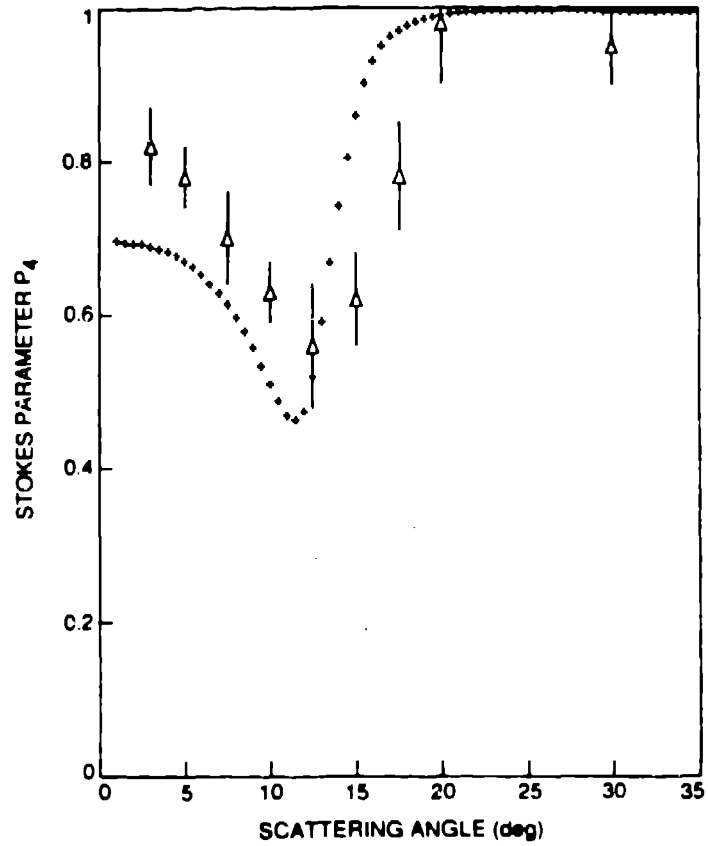

Fig. 4 Model calculations and experimental data[9] for excitation of Kr $5s'$ $[1/2]_1^0$ $^1P_1$ at 60 eV.

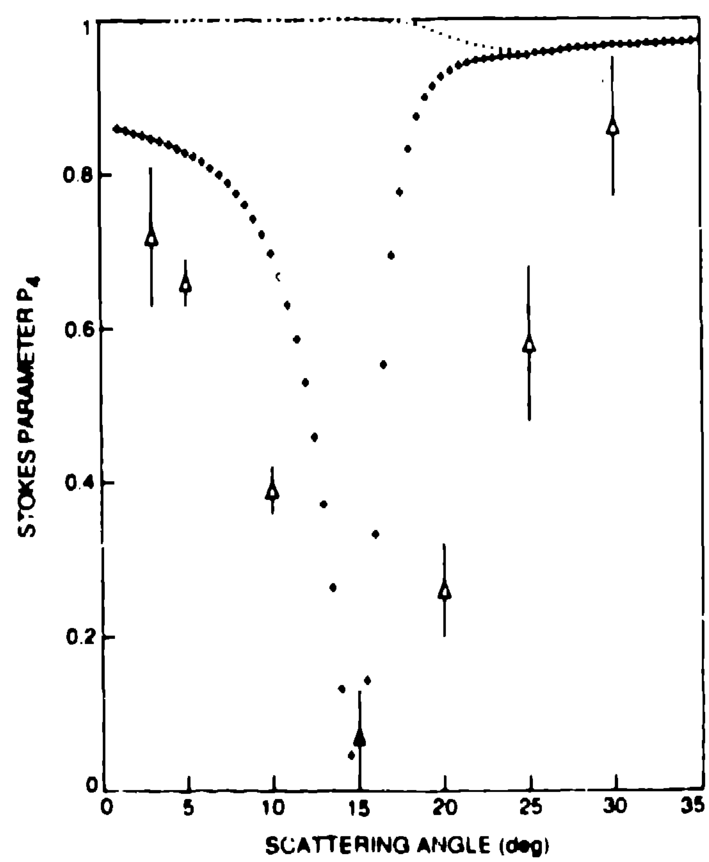

Fig. 5 Model calculations and experimental data[9] for excitation of Xe $6s[3/2]_1^0$ $^3P_1$ at 30 eV.

scattering at the laboratory frame origin, P4, in the case of Kr, is expected to be unity over the range of scattering angles studied, while, in the case of Xe, the dotted line in Fig. 5 shows the predicted behavior of P4.

These results will be discussed in more detail in a forthcoming publication[11].

Conclusions

Results of our modelling effort imply that measurements of EICP using experimental geometries in which the photon incidence vector (either laser photon in the superelastic scattering experiment or detected photon in the polarization correlation experiment) lies in the nominal scattering plane can be subject to large uncertainties. The influence of an extended scattering volume can produce severe distortion in the measured modulation depth, η, or Stokes parameter, P4, which can be misinterpreted as arising from spin- orbit coupling effects. The severity and location (in scattering angle) of this distortion is intimately tied to the behavior of the P_ℓ^+ and γ parameters (or, equivalently, the λ and χ parameters).

Acknowledgements

This work was supported by the National Science Foundation, the National Aeronautics and Space Administration and the U. S. Department of Energy.

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